



An analytical model for alkaline membrane direct methanol fuel cell



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ABSTRACT

In this study, a multiphase analytical model is developed for alkaline anion exchange membrane direct methanol fuel cell (AAEM-DMFC). The model prediction agrees with experimental data reasonably. Modeling results show that the methanol feed concentration, operating temperature and membrane thickness are the three factors that most significantly affect the cell performance. The effect of reactant flow rate is insignificant in high flow rate range, and this effect enhances when the flow rates are low. In low current density range, the cell shows better performance with lower methanol feed concentrations, while this trend reverses in high current density range. A similar trend is also found for the operating temperature. A thinner membrane leads to a higher methanol crossover; however, it yields better performance in mid and high current density range. Water is mass transfer limited once membrane thickness is high enough, resulting in the decrease of limiting current density. The carbon dioxide bubbles produced in anode are removed faster at higher operating temperatures. When the anode of the cell faces up, the best performance can be achieved. Inclining the cell leads to lower cell performance, and the performance degradation becomes more significant with larger inclining angles.

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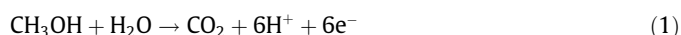
1. Introduction

There are active researches in the past two decades focusing on the direct methanol fuel cell (DMFC), which is considered as an ideal candidate for portable electronic applications due to its convenient fuel storage and handling, high energy density and low operating temperature [1,2].

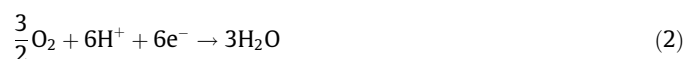
In the early 1950s, Palve first demonstrated that methanol can act as a fuel in aqueous electrolytes [3], since the development of solid polymer electrolytes (mainly perfluorinated cation exchange membranes), proton exchange membranes (PEMs) have been widely used for DMFC applications. Nowadays, PEM-DMFC still faces two major obstacles, namely, the relatively sluggish kinetics at the anode and methanol crossover from anode to cathode [4], which hinder its commercialization.

The reactions occurring in the electrodes of a PEM-DMFC are:

Anode oxidation:



Cathode reduction:



Overall reaction:



In the electrode reaction at the PEM-DMFC anode, by consuming 1 mol of methanol, 6 mol of hydrogen ion and 6 mol of electron are produced; on the other hand, 1 mol of water needs to be consumed (needs effective delivery of water to the electrode), and 1 mol of CO₂ is produced (needs effective removal of CO₂ from the electrode), which may make the mass transport difficult. In addition, the methanol crossover from anode to cathode may also cause parasitic consumption of fuel, flooding of cathode and other problems lowering the performance [5].

Many mathematical models have been developed to study the heat and mass phenomena coupled with the electrochemical reactions [6–12]. Among these models, considerable attention has been devoted to analytical model, because it provides qualitative insights into the transport processes in the fuel cells based on fundamental considerations and appropriate assumptions [10–12]. For example, Rosenthal et al. [11] developed a one-dimensional analytical model for PEM-DMFC to study the effect of a variety of operating parameters (such as methanol feed concentration and

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Nomenclature

<i>a</i>	activity	ϕ	average gas relative humidity
<i>A</i>	cell geometric area (m ²)	<i>Subscripts and superscripts</i>	
<i>C</i>	molar concentration (mol m ⁻³); gas constant; viscous drag coefficient	0	intrinsic value, standard condition, reference
<i>d</i>	diameter (m)	<i>a</i>	anode
<i>D</i>	mass diffusivity (m ² s ⁻¹)	<i>act</i>	activation process
<i>E</i>	potential (V); effective activation energy (J mol ⁻¹)	AAEM	alkaline anion exchange membrane
<i>EW</i>	equivalent weight of the membrane (kg mol ⁻¹)	ACL	anode catalyst layer
<i>F</i>	Faraday's constant (96485.0 C mol ⁻¹); force (N)	ADL	anode diffusion layer
<i>g</i>	acceleration of gravity (m s ⁻²)	AFC	anode flow channel
<i>i</i>	current density (A m ⁻²)	<i>bub</i>	bubble
<i>k</i>	Henry's constant	<i>buo</i>	buoyancy
<i>K</i>	permeability (m ²)	<i>c</i>	cathode
<i>M</i>	molecular weight (kg mol ⁻¹)	<i>cap</i>	capillary
<i>n</i>	stoichiometric coefficient of electrons in electrode reaction; molar number and pore number (mol)	<i>cell</i>	fuel cell
<i>N</i>	Flux (mol m ⁻² s ⁻¹)	<i>cross</i>	crossover
<i>P</i>	pressure (Pa); partial pressure of gas with zero dimension	CC	concentration vs. concentration
<i>R</i>	universal gas constant (8.314 J K ⁻¹ mol ⁻¹); cell resistivity (Ω m ²); radius (m)	CCL	cathode catalyst layer
<i>Re</i>	Reynolds number	CDL	cathode diffusion layer
<i>s</i>	volume fraction; entropy (J mol ⁻¹ K ⁻¹)	CFC	cathode flow channel
<i>Sh</i>	Sherwood number	CO ₂	carbon dioxide
<i>T</i>	temperature (K)	<i>diss</i>	dissolved
<i>v</i>	velocity (m s ⁻¹)	<i>eff</i>	effective
<i>V</i>	voltage (V); volume (m ³); partial molar volume (m ³ mol ⁻¹)	<i>g</i>	gas
<i>x</i>	position or coordinate (m)	<i>h</i>	hydraulic
<i>Greek symbols</i>		<i>H</i>	Henry's law
Λ	volumetric flow rate (m ³ s ⁻¹)	H ₂ O	water
α	kinetic transfer coefficient	<i>in</i>	inlet
β	cell inclining angle (°)	<i>lq</i>	liquid water
δ	thickness (m)	<i>m</i>	membrane
ε	porosity	<i>mol</i>	molecular
η	voltage loss (V)	Me	methanol
θ	contact angle (°)	<i>ohm</i>	ohmic
λ	water content in ionomer	<i>out</i>	outlet
μ	dynamic viscosity (kg m ⁻¹ s ⁻¹)	O ₂	oxygen
ζ	electro-osmotic drag coefficient (H ₂ O per OH ⁻)	<i>pore</i>	pore
ρ	density (kg m ⁻³)	<i>ref</i>	reference
σ	membrane conductivity (S m ⁻¹); surface tension coefficient (N m ⁻¹)	<i>rs</i>	electrode reaction
		<i>sat</i>	saturation
		<i>tran</i>	transport process
		<i>vis</i>	viscous resistance
		<i>w</i>	water
		<i>wv</i>	water vapor
		ϕ	potential

operating temperature) on the cell performance. Their results suggest that increasing the methanol feed concentration results in higher methanol crossover and lower open circuit voltage (OCV). At higher methanol feed concentration, the OCV is not a monotonic function of the operating temperature, which only holds at lower methanol feed concentrations. Another one-dimensional analytical model was developed for liquid-feed PEM-DMFC by Kareemulla and Jayanti [12]. They used a multi-step reaction mechanism to describe the methanol electrochemical reaction at the anode, with Stefan–Maxwell equations to describe the multi-component diffusion in the cathode. Their results suggested that at high methanol feed concentration, oxygen depletes on the cathode side due to the excessive methanol crossover, which results in substantial mass transport loss. Although active research has been conducted in the PEM-DMFC theoretical modeling and fundamental understanding of heat and mass transfer, the high cost of PEMs, platinum-based catalysts, substantial fuel crossover and sluggish reaction kinetics appear to be unavoidable, which limit its development and application.

In recent years, researchers are turning their interest toward alkaline anion exchange membrane direct methanol fuel cell (AAEM-DMFC). It is known that the electrochemical reaction kinetics in alkaline electrolyte is faster than that in acidic media [13]. In fact, one of the first DMFCs developed by Justi and Winsel in 1955 was operated with alkaline aqueous electrolytes [14]. However, the research on alkaline DMFC had to be scaled down in 1980s due to the electrolyte poisoning caused by CO₂ production of methanol oxidation and from air, which limited its application [15].

In order to reduce the formation of carbonate in the electrolyte caused by CO₂, researchers solidified the alkaline media, namely, replaced the liquid electrolyte with solid electrolyte, so that air might be used in AAEM-DMFC. This direction can be reflected by an impressive growth in publications related to AAEMs recent years [16–20]. Xiong et al. [16] presented a new way to prepare AAEMs based on polyvinyl alcohol (PVA). They grafted quaternary ammonium groups as charge carriers onto the PVA backbone by using (2,3- epoxypropyl)trimethylammonium chloride. The conductivities of the cross-linked quaternized-PVA were in the order

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